

Quantum transport through a molecular level: a scattering states numerical renormalisation group study

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We use the scattering states numerical renormalization group (SNRG) approach to calculate the current $I(V)$ through a single molecular level coupled to a local molecular phonon. The suppression of I for asymmetric junctions with increasing electron-phonon coupling, the hallmark of the Franck-Condon blockade, is discussed. We compare the SNRG currents with recently published data obtained by an iterative summation of path integrals approach (ISPI). Our results excellently agree with the ISPI currents for small and intermediate voltages. In the linear response regime $I(V)$ approaches the current calculated from the equilibrium spectral function. We also present the temperature and voltage evolution of the non-equilibrium spectral functions for a particle-hole asymmetric junction with symmetric coupling to the lead.

I. INTRODUCTION

In the quest for size-reduced and possible low-power consuming electronic devices, the proposal [1] of using molecular junctions for electronics has sparked a large interest in understanding the influence of molecular vibrational modes onto the electron charge transfer through a molecule. The non-linear current through a molecule device can be controlled by a capacitively coupled external gate [2, 3]. Interestingly, hysteretic behavior of the $I(V)$ curves [4] has been reported in several experiments when sweeping the voltage with a finite rate. However, the observed hysteresis are non-universal and depend on the sweeping rate. For infinitesimally slow sweeping the effect vanishes. In some cases a sudden drop of the current has been observed with increasing bias voltage [2] which translates into a negative differential conductance. This all has been accounted to configurational changes of the molecule emphasizing the importance of vibrational couplings in such devices.

Many experimental facts have been gathered in the last two decades but there is still a lack of an accurate theoretical description of all the reported phenomena. An excellent review [5] by Galperin et al. comprehensively summarizes the different theoretical approaches and experimental findings. Single molecular transistors (SMT) promise to offer some advantages over their semiconductor based counterparts [6]. Both types of single-electron transistors can be controlled by a capacitively coupled external gate [2, 3, 6]. The molecular energy scales, however, are larger in SMTs and reproducibly defined by the chemistry of the molecule. In addition, the coupling to vibrational modes enlarges the parameter space and different physics such a phonon-assisted tunneling, Frank-Condon blockade or the appearance of inelastic steps in the $I(V)$ curve can be observed.

The theoretical description of such molecular junctions only include those molecular levels and vibrationals modes relevant for the quantum transport. The simplest model proposed [5, 7, 8] comprise a single level coupled to a local Holstein phonon. Typically rate equations [9] or lowest order Keldysh-Green function approaches

[5, 10, 11] have been applied to this problem [7]. Recently, the iterative path-integral approach (ISPI) [12] has also been successfully applied [13] to calculate quantum transport for moderate and high temperatures compared to the charge-transfer rate Γ_0 .

The equilibrium physics of two extreme limits have been well understood in a model containing only a single vibrational mode [5, 9, 14]. In the adiabatic limit, the phonon frequency is the smallest energy scale of the problem and a small electron-phonon coupling yields a reduction of the phonon frequency by particle-hole excitations. This limit has been pioneered by Caroli et al. [15] in the context of tunnel junctions and applied to molecular junctions [8].

In the opposite limit, for very small tunneling rates t_α one starts from the exact solution of the local problem by applying a Lang-Firsov transformation [16]. A displaced phonon with an unrenormalized phonon frequency ω_0 and a polaron with a shifted single-particle energy is formed locally. In this anti-adiabatic limit, the strong electron-phonon coupling yields a polaronic shift of the single-particle level and an exponential suppression of tunneling rate related to the Franck-Condon blockade [5, 9, 13, 17].

Wilson's numerical renormalization group (NRG) approach [18, 19] has been adapted to the Holstein model in equilibrium [20]. A comprehensive study [14] has demonstrated the power of this non-perturbative approach to reveal the interplay between the different energy scales of the problem in the crossover regime. In this article we review the extension [21] of the approach to steady-state currents by applying the scattering-states NRG (SNRG) [22–24] to the spinless Anderson-Holstein model.

II. THEORY OF QUANTUM TRANSPORT THROUGH MOLECULAR JUNCTION

A. Model

In molecular electronics experiments [2, 3], a complex organic molecule is contacted by two conducting

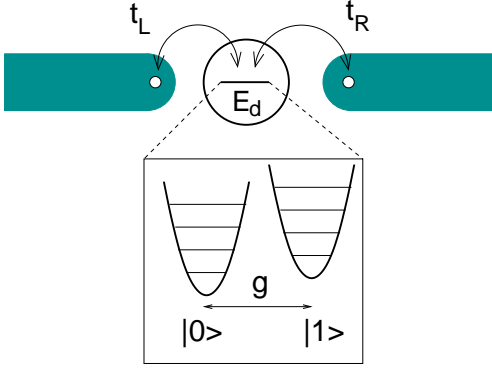


FIG. 1. Minimal model of a molecule consisting of a single active molecular level at energy E_d coupled to two leads with tunneling matrix elements t_L and t_R . Depending on the local charge configuration $|0\rangle$ or $|1\rangle$, the ground state of a vibrational degree of freedom is shifted. The relative displacement between the two configuration is given by the dimensionless electron phonon coupling $g = \lambda_{ph}/\omega_0$. The phononic excitations for a fixed charge are multiples of the oscillator energy ω_0 .

leads. We have modeled these leads as two symmetric featureless free electron gases since the mean-free path in the leads is large compared to the spatial dimensions of the device. In general, the molecule can contain several molecular orbitals which are actively participating in the quantum transport. Furthermore, the internal vibrational modes of the molecule are influenced by charging and discharging of the molecule.

Here we consider the most minimalistic model for quantum transport through a molecule [5, 9, 13] and follow the notation of Ref. [21]. This widely used Hamiltonian [5, 13, 14] is defined as

$$\begin{aligned}
 H = & E_d d^\dagger d + \omega_0 b^\dagger b + \lambda_{ph} (b^\dagger + b) \left(\hat{n}_d - \frac{1}{2} \right) \\
 & + \sum_{\alpha=L,R} \sum_k \varepsilon_{k\alpha} c_{k\alpha}^\dagger c_{k\alpha} \\
 & + \sum_{\alpha=L,R} \frac{t_\alpha}{\sqrt{N}} \sum_k (d^\dagger c_{k\alpha} + c_{k\alpha}^\dagger d)
 \end{aligned} \quad (1)$$

where $d(d^\dagger)$ annihilates(creates) an electron on the device with energy E_d , and $c_{k\alpha}^\dagger$ creates an electron in the lead α with energy $\varepsilon_{k\alpha}$. The local charge-transfer rate to each lead α is given by $\Gamma_\alpha = \pi t_\alpha^2 \rho_\alpha(0)$, where $\rho_\alpha(\omega)$ is the density of states of lead α . In order to focus only on the influence of the electron-phonon interaction onto the quantum transport, the spin degree of freedom is neglected in order to avoid obstruction of the competition between spin-flip scattering through the device and polaron formation on the device.

The model comprise a single active molecular level - all others are energetically well separated - whose charge density is coupled to a local Holstein phonon stemming from the dominating vibrational mode of the molecule.

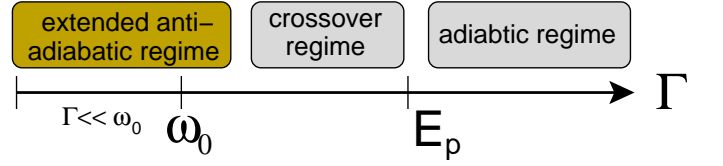


FIG. 2. The different regimes as function of the charge transfer rate Γ_0 . The crossover from extended anti-adiabatic regime is reached when $\Gamma_{\text{eff}} \approx e^{-g^2} \Gamma_0 \approx \omega_0$ and extends to $\Gamma \approx E_p = g^2 \omega_0$.

In real materials, band features are important but only influence the single-particle properties which can be accounted for in a frequency dependent charge transfer rate $\Gamma_\alpha(\omega)$ which we treat as a constant for simplicity in our simulations.

The spinless Anderson-Holstein model is schematically depicted in Fig. 1. Depending on the local charge configuration, the local harmonic oscillator is displaced and the dimensionless distance between the two ground states is given by $g = \lambda_{ph}/\omega_0$. For modeling realistic situations, the restriction to a single phonon and a single electronic level must be lifted. In spite of a lot of theoretical progress [5] this model has only been accurately solved in equilibrium [14, 20, 25], while its non-equilibrium dynamics has only been perturbatively investigated in lowest order of the coupling constants [5].

The local Hamiltonian is given by the first line in (1) and can be solved exactly using the Lang-Firsov transformation [16, 26]. This local solution consists of a local polaron decoupled from a shifted harmonic oscillator. The corresponding polaronic energy gain is given by $E_p = \lambda_{ph}^2/\omega_0 = g^2 \omega_0$.

Coupling this local degrees of freedom to the two leads defines two competing regimes depicted in Fig. 2. For $E_p, \omega_0 \ll \Gamma_0 = \Gamma_L + \Gamma_R$, the phonon dynamics is slow and can be treated perturbatively in this adiabatic regime. $E_p, \omega_0 \gg \Gamma_0$ defines the opposite limit: in this anti-adiabatic regime charge fluctuations are suppressed, the electron moves slow and the phonon defines the large energy scale. The anti-adiabatic regime is relevant for molecular junctions since the tunneling coupling of a molecule to the leads is usually small compared to the intrinsic energy scales of the molecule. After the Lang-Firsov transformation, the tunneling term acquires an additional factor $\exp[g(b^\dagger - b)]$ whose physical meaning is stripping the original electron content from the locally formed polaron. If $\omega_0 \gg \Gamma$, the local phonon remains in its ground states which yields an exponential suppression of the tunneling coupling and $\Gamma \rightarrow \Gamma_{\text{eff}} \approx \Gamma_0 e^{-g^2}$. In a particle-hole asymmetric junction, this leads to a Franck-Condon suppression of the current for small bias voltage: The system reacts with a dynamical suppression of the tunneling rate to avoid the reorganization of the nuclear positions of the molecule.

B. Scattering-states numerical renormalization group

The scattering-states numerical renormalization group (SNRG) approach is based on the steady-state density operator $\rho(\mu_L, \mu_R)$ for a current carrying ensemble [27] coupled to two baths at different chemical potentials μ_α . Hershfield has shown [27] that this operator has a Boltzmannian form

$$\hat{\rho}(\mu_L, \mu_R) = \frac{1}{Z} e^{-\beta(H-Y)} \quad (2)$$

where Z is the partition function and $\mu\hat{N}$ is replaced by the Y -operator. This Y -operator is in general unknown for an arbitrary fully interacting Hamiltonian.

For a non-interacting problem, however, the corresponding Y_0 -operator is given in terms of the Lippmann-Schwinger scattering states with energy ε of left-moving or right moving single-particle scattering state created by $\gamma_\alpha^\dagger(\varepsilon)$,

$$Y_0 = \sum_\alpha \mu_\alpha \int d\varepsilon \gamma_{\varepsilon\alpha}^\dagger \gamma_{\varepsilon\alpha} \quad (3)$$

where $\{\gamma_{\varepsilon\alpha}^\dagger, \gamma_{\varepsilon'\alpha'}^\dagger\} = \delta_{\alpha\alpha'}\delta(\varepsilon - \varepsilon')$. Note that the applied source-drain voltage $eV = \mu_L - \mu_R$.

In the SNRG [22, 23] we have circumvented the unknown Y by the following procedure.

First, we realise that we can discretize the energy dependent scattering states on a logarithmic energy mesh identically as in the standard NRG [19, 22] such that we obtain a two-band model comprising of a left-mover and right-mover band. (Below, we will comment on the analytical form of the scattering states.) Then we perform a standard NRG using $K_0 = H(\lambda_{ph} = 0) - \hat{Y}_0$.

Knowing the analytical form of the non-equilibrium density operator $\hat{\rho}_0(V) = \exp(-\beta K_0)/Z_0$, we can discretize scattering states on a logarithmic energy mesh identically to the standard NRG [19, 22] and perform an NRG using $K_0 = H(\lambda_{ph} = 0) - \hat{Y}_0$. The density operator $\hat{\rho}_0(V)$ contains all information about the current carrying steady-state for the Hamiltonian $H_0 = H(\lambda_{ph} = 0)$ [27].

Starting at time $t = 0$, we let the non-interacting system propagate with respect to the full Hamiltonian $H_f = H(\lambda_{ph} > 0)$: The density operator $\hat{\rho}(t)$ progresses as $\hat{\rho}(t) = \exp(-iH_f t)\rho_0 \exp(iH_f t)$. Since we quench the system only locally, we can assume $\hat{\rho}(t)$ reaches a steady-state at $t \rightarrow \infty$ independent of initial condition for an infinitely large system: all bath correlation functions decay for infinitely long times. The finite size oscillations always present in the NRG calculation [22, 28–30] are projected out by defining the time-averaged density operator [22, 31]

$$\hat{\rho}_\infty = \lim_{T \rightarrow \infty} \frac{1}{T} \int_0^T dt \hat{\rho}(t). \quad (4)$$

Consequently, only density matrix elements diagonal in energy contribute to the steady-state in accordance with

the condition $[H_f, \hat{\rho}_\infty] = 0$. Even though the Y -operator remains unknown, we explicitly construct a numerical representation of the non-equilibrium density matrix using the time-dependent NRG [28, 29]. In a last step, we calculate local steady-state retarded Green function

$$G_{d,d^\dagger}^r(t) = -i\text{Tr} [\hat{\rho}_\infty \{d(t), d^\dagger\}] \Theta(t), \quad (5)$$

where $d(t) = e^{iH_f t} d e^{-iH_f t}$ and $\hat{\rho}_\infty$ has been defined in Eq. (4). The approach is based on an extension for equilibrium Green functions [32] and its technical details are found in Ref. 33.

It has been show [27, 34, 35] that for the model investigated here, the current is given by the by a generalized Landauer formula

$$I(V) = \frac{G_0}{e} \int_{-\infty}^{\infty} d\omega [f_R(\omega) - f_L(\omega)] \Gamma_0 \pi \rho_d(\omega, V) \quad (6)$$

where $f_\alpha(\omega) = f(\omega - \mu_\alpha)$ and the steady-state spectral function $\pi \rho_d(\omega, V) = \Im m[G_{d,d^\dagger}^r(\omega - i0^+, V)]/\pi$ is obtain from the Fourier transformed retarded Green function Eq. (5). The prefactor

$$G_0 = \frac{e^2}{h} \frac{4\Gamma_L \Gamma_R}{\Gamma_0^2} \quad (7)$$

contains the leading asymmetry factors of the junction and reaches the universal conductance quantum e^2/h for a symmetric junction, i. e. $\Gamma_L = \Gamma_R$. G_0 can be expressed as $G_0 = (e^2/h)(4R/(1+R)^2)$ using the definition of the coupling asymmetry ratio $R = \Gamma_L/\Gamma_R$.

C. Single-particle scattering states

In the absence of the electron-phonon coupling, the Hamiltonian (1) can be solved exactly in terms of single-particle Lippmann-Schwinger scattering states [22, 27, 35, 36],

$$H_0 = \sum_\alpha \int d\varepsilon \varepsilon \gamma_{\varepsilon\alpha}^\dagger \gamma_{\varepsilon\alpha} \quad (8)$$

where

$$\gamma_\alpha^\dagger = c_{\varepsilon\alpha}^\dagger + t_\alpha \sqrt{\rho_\alpha(\varepsilon)} G_{0\sigma}^r(\varepsilon + i\delta) \times \left[d^\dagger + \sum_{\alpha'} \int d\varepsilon' \frac{V_{\alpha'} \sqrt{\rho_{\alpha'}(\varepsilon')}}{\varepsilon + i\delta - \varepsilon'} c_{\varepsilon'\alpha'}^\dagger \right] \quad (9)$$

and the local resonant level Green function

$$G_0^r(z) = [z - E_d - \Delta(z)]^{-1} \quad (10)$$

$$\Delta(\omega - i\delta) = \sum_\alpha t_\alpha^2 \int d\varepsilon \frac{\rho_\alpha(\varepsilon)}{\omega - \varepsilon} \quad (11)$$

enters as one of the expansion coefficients. $\rho_\alpha(\varepsilon)$ denotes the density of states of the individual leads and will be takes as equal and featureless in the following. The small imaginary part $i\delta$ is required for regularization in the

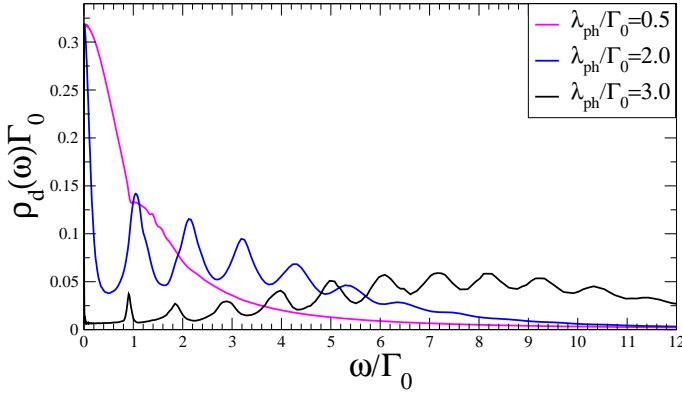


FIG. 3. Equilibrium spectral function of the particle-hole symmetric junction, i. e. $E_d = 0$, and $\omega_0/\Gamma_0 = 1$ for three different electron-phonon coupling strength $\lambda_{ph}/\Gamma_0 = 0.5, 2, 3$.

transition from the discrete k summation in Eq. (1) to the continuum limit an caused the time-reversal symmetry breaking.

Note that the d -orbital has been included into the scattering states. By inverting the unitary transformation, we can expand the local d -orbital in left-moving and right-moving scattering states.

$$d^\dagger = r_R d_R^\dagger + r_L d_L^\dagger \quad (12)$$

$$d_\alpha^\dagger = \bar{t} \int d\varepsilon \sqrt{\rho(\varepsilon)} [G_0^r(\varepsilon + i\delta)]^* \gamma_{\varepsilon\alpha}^\dagger \quad (13)$$

where we defined $\bar{t} = \sqrt{t_L^2 + t_R^2}$ and have used $r_\alpha = t_\alpha/\bar{t}$. The expansion coefficients in Eq. (13) contain the retarded Green function $G_0^r(\varepsilon + i\delta)$ which we separate in modulus and phase

$$[G_0^r(\varepsilon + i\delta)] = |G_0^r(\varepsilon + i\delta)| e^{-i\Phi(\varepsilon)}. \quad (14)$$

This phase is absorbed into the new scattering states $\gamma_{\varepsilon\alpha}^\dagger \rightarrow \tilde{\gamma}_{\varepsilon\alpha}^\dagger = \gamma_{\varepsilon\alpha}^\dagger e^{i\Phi(\varepsilon)}$ by a local gauge transformation. In the wide band limit, i.e. $D \gg \Gamma_0$, the effective DOS $\tilde{\rho}(\varepsilon) = [\bar{t} \sqrt{\rho(\varepsilon)} |G_0^r(\varepsilon - i\delta)|]^2$ is normalized,

$$\int d\varepsilon \tilde{\rho}(\varepsilon) = \int d\varepsilon \left[\bar{V} \sqrt{\rho(\varepsilon)} |G_0^r(\varepsilon - i\delta)| \right]^2 = 1, \quad (15)$$

and d_α^\dagger is used as a starting vector for the Householder transformation [18, 19] for constructing the discretized Wilson chain. Although the physical contained of the Wilson chain sites are different to the standard NRG [18, 19] the analytical form is preserved [22]. Since the local gauge transformation has to applied to the local current operator, the current flow is related to $\sin(\Phi(\varepsilon))$ of the energy dependent scattering phase.

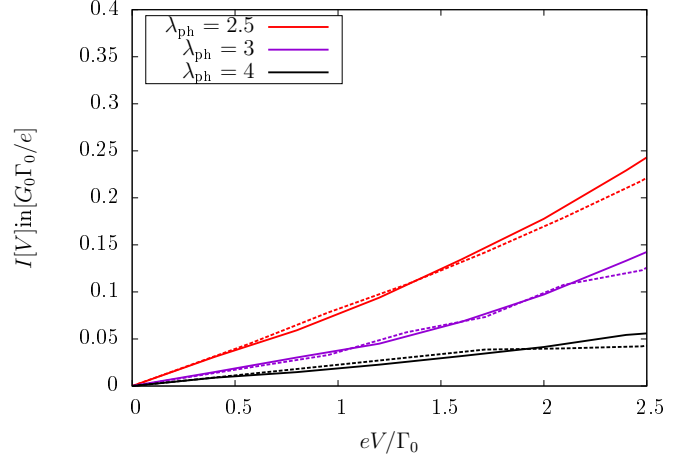


FIG. 4. The evolution of $I(V)$ curves from medium to strong electron-phonon coupling for $T/\Gamma_0 = 0.2$ for a fixed phonon frequency $\omega_0/\Gamma_0 = 2$, a symmetric junction $\Gamma_L = \Gamma_R$ and level position in H_m , i. e. $\varepsilon = E_d + \lambda_{ph}^2/\omega_0 = 0$. A comparison between the SNRG data (straight lines) and the ISPI data (dotted lines), taken with permission from Ref. [13], is shown.

III. RESULTS

A. Equilibrium spectral function

To set the stage for the non-equilibrium steady state currents, we show the evolution of the equilibrium spectra function $\rho_d(\omega)$ for three different ratios λ_{ph}/Γ in Fig. 3. While $\lambda_{ph}/\Gamma = 0.5$ lies in the adiabatic regime, the two others are represents the anti-adiabatic regime. For $\lambda_{ph}/\Gamma = 0.5$, we find a kink in the spectral function at ω_0 where strong electron-phonon scattering sets in. For $\lambda_{ph}/\Gamma = 2$ we observe already very pronounced phonon-replicas with a reduced width. Increasing λ_{ph}/Γ further yields to a substantial shift of spectral weight from the resonance at $\omega = 0$ to larger frequencies: a careful analysis shows [14, 21] that the peak of the envelope function is related to the effective Coulomb repulsion between the d -electron and the conduction band electrons which can be derived analytically for $t_\alpha \rightarrow 0$ using a Schrieffer-Wulff transformation [14].

B. Steady-state currents

Recently, a numerical approach based on the iterative summation of path integrals (ISPI) [12] has been applied [13] to the model defined in Eq. (1). Since it requires a fast decay of the memory kernel for the discretized iterative summation of the path integral, it is restricted to moderate and high temperatures for large electron-phonon couplings. In this section, we will provide a comparison of the ISPI with the SNRG using the published ISPI data of Ref. [13].

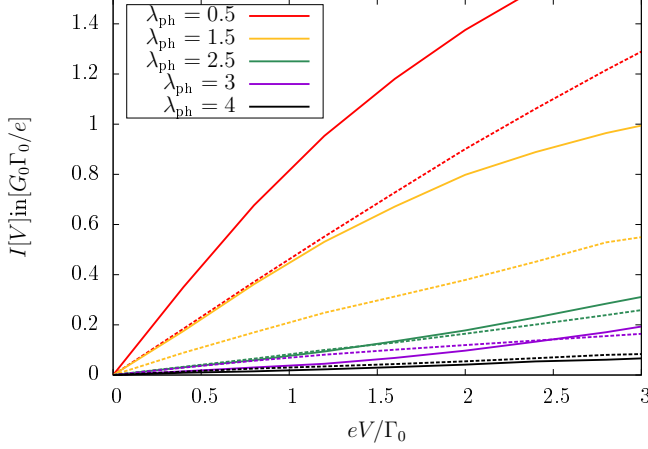


FIG. 5. The evolution of $I(V)$ curves from weak to strong electron-phonon coupling for $T/\Gamma_0 = 1$ (dashed lines) and $T/\Gamma_0 = 0.2$ (straight lines) calculated with the SNRG. We have set $\omega_0/\Gamma_0 = 2$, a symmetric junction $\Gamma_L = \Gamma_R$ and $\varepsilon = E_d + \lambda_{ph}^2/\omega_0 = 0$.

It is straight forward to show that local Hamiltonian

$$H_m = \varepsilon d^\dagger d + \omega_0 b^\dagger b + \lambda_{ph}(b^\dagger + b)\hat{n}_d \quad (16)$$

commonly used in the literature [7, 13] yield the same dynamics as the first three terms in Eq. (1) after identifying $\varepsilon = E_d + g^2\omega_0$ and performing a linear shift of the bosonic operators [14, 21].

Figure 4 shows the evolution of the current for a symmetric junction $\Gamma_L/\Gamma_R = 1$ from medium to strong coupling at $T/\Gamma_0 = 0.2$, $\varepsilon = 0$ and $\omega_0/\Gamma_0 = 2$. The overall agreement between the SNRG data (solid lines) and the ISPI approach (dotted lines) is remarkable up to $eV \approx 2\Gamma_0$ after which the small deviations become more pronounced: The SNRG current slightly exceeds the ISPI data for large voltages. Since both approaches, the ISPI and the SNRG, rely on discretisation of a continuum, we believe that the origin of these deviations are related to the different discretisation errors inherent in both methods.

The temperature dependency of the SNRG $I(V)$ curves are shown in Fig. 5 for two different temperatures. We combine the data of Fig. 4 for $T/\Gamma_0 = 0.2$ (straight lines) with the $I(V)$ for the same parameters but calculated at $T/\Gamma_0 = 1$ (dashed lines). In the limit $T \rightarrow \infty$ the currents must vanish: In this high temperature limit all left and right moving scattering states are equally occupied leading to zero net current as predicted by Eq. (6). For the electron-phonon couplings $\lambda_{ph}/\Gamma_0 = 0.5, 1.5, 2.5$, we clearly observe a decrease of the current with increasing temperature.

Above $\lambda_{ph}/\Gamma_0 = 3$, we observe a qualitative change of the behavior: the low temperature current is smaller than its high temperature counterpart: an indication of the Franck-Condon blockade in the quantum transport.

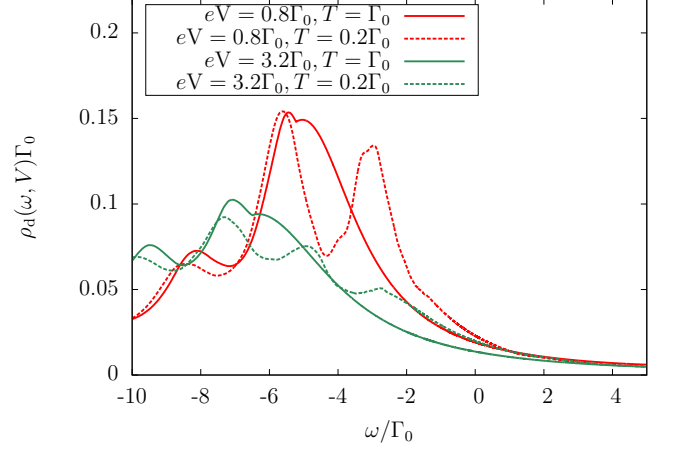


FIG. 6. SNRG nonequilibrium spectral functions for two different voltages and two temperatures. We have set $\omega_0/\Gamma_0 = 2$, $\lambda_{ph}/\Gamma_0 = 3$ and $\varepsilon = E_d + \lambda_{ph}^2/\omega_0 = 0$.

There are two contributions changing the current according to Eq. (6) for a fixed voltage when raising the temperature. Firstly, the Fermi window becomes flatter and broader, and the high energy parts of the spectral function contribute stronger. In addition, the spectral function shows a significant temperature and voltage dependency with increasing electron-phonon coupling.

To illustrate this points, a comparison of nonequilibrium spectral functions with $\lambda_{ph} = 3$ is depicted in Fig. 6 for two different temperatures and two different bias voltages. For $eV = 0.8\Gamma_0$ an increase of temperature leads to a suppression of the phonon side peak at $\omega/\Gamma_0 = -3$.

At the same time the peak at $\omega/\Gamma_0 = -5$ is broadened and contributes more weight to the integral due to the broadened Fermi window, leading to an increase of the current with increasing temperature. At a voltage of $eV = 3.2\Gamma_0$ the decrease of the spectral weight at $\omega/\Gamma_0 = -3$ is not compensated within the Fermi window contributing to the current integral leading to a decrease of the current with increasing temperatures. Therefore, we observe a crossover between an increase of current at small voltages to an decrease of current at large voltages with increasing temperatures.

In contrary, the difference between the I-V curves of $T/\Gamma_0 = 1$ and $T/\Gamma_0 = 0.2$ are large at low phonon-couplings $\lambda_{ph}/\Gamma_0 = 0.5, 1.5$. In this perturbative regime, the spectral function is only very weakly temperature dependent, and, therefore, the change of currents is only related to the temperature dependence of the Fermi functions in Eq. (6).

C. Linear response regime

In order to explicit reveal the influence of the voltage dependency of the spectral function on the current $I(V)$,

we compare the SNRG curves with the current calculated with the equilibrium spectral function $\rho_d(\omega, V) = \rho_d(\omega)$ in Eq. (6) neglecting its voltage dependency. The latter becomes asymptotically exact for $V \rightarrow 0$, defining the linear response regime. Deviations from these curves are caused by the voltage dependency of the true non-equilibrium spectral function. The results are depicted in Fig. 7 using the SNRG data of Fig. 5 for $T = \Gamma_0$. The SNRG curves (straight line) coincide with the equilibrium calculation (dashed line) in the linear response regime, i. e. $|eV| \ll \Gamma_0$. However, the larger the electron-phonon coupling, the smaller the validity range of the linear response regime. Already at very small finite voltages, we observe deviations from the $I(V)$ generated by the equilibrium $\rho_d(\omega)$. The excellent agreement between the ISPI and the SNRG for $\lambda_{ph}/\Gamma_0 = 4$ results in the small voltage regime – see Fig. 4 – clearly demonstrates that the SNRG correctly accounts for the bias dependence of the spectral function.

IV. CONCLUSION

We have applied the scattering states numerical renormalization group approach to the charge-transport through a symmetric molecular junction. Since we have focused on the influence of a vibronic mode on the transport, we have restricted ourselves to the investigation of the spinless Anderson-Holstein Model. We have started with a brief review of the different regimes of the model and have connected them to the polaronic energy shift E_p .

To set the stage for the non-equilibrium steady state currents we have performed equilibrium calculations and have analysed the equilibrium spectral functions in the different regimes. We have demonstrated the Franck-Condon blockade in the $I(V)$ curves found in the particle-hole asymmetric case: the current is increasingly suppressed with increasing electron-phonon coupling.

We have shown the temperature evolution of the $I(V)$ for two different moderate temperatures to make contact to the ISPI approach [13]. While the ISPI is limited to large temperatures due to the discretization of the memory kernel, the SNRG can access arbitrarily low temperatures, the quantum coherence dominates the transport properties at low temperatures.

At small voltages and strong electron-phonon coupling $\lambda_{ph}/\Gamma_0 > 2.5$ the shape change of the non-equilibrium spectral function leads to a suppression of the current

when the temperature is increased. The temperature dependency of the current is governed by the Fermi-functions of the lead for large voltages or small couplings λ_{ph} . We have shown that our non-equilibrium currents approach the linear response regime for small voltages in which the voltage dependency of the spectral function can be neglected. With increasing λ_{ph} , however, the validity radius of the linear response regime becomes very small.

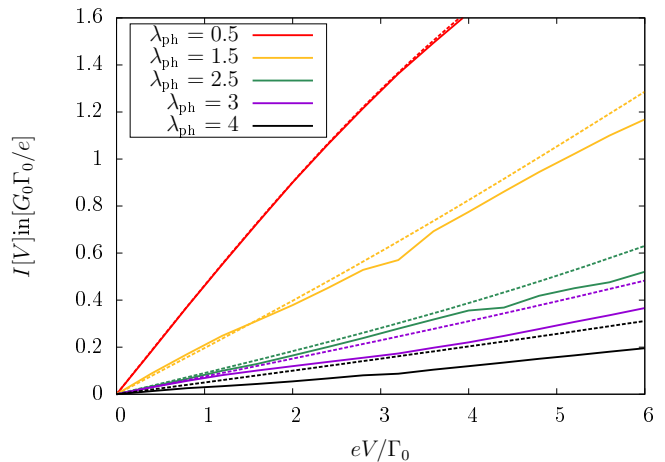


FIG. 7. The evolution of $I(V)$ curves from weak to strong electron-phonon coupling for the same parameters as in fig. 4 but $T/\Gamma_0 = 1$. The straight lines are SNRG nonequilibrium results and the dashed lines are $I(V)$ curves where we set $\rho_d(\omega, V) = \rho_d(\omega)$ in Eq. 6.

V. ACKNOWLEDGMENTS

This paper is dedicated to the memory of Avi Schiller. He was not only a dear friend but a collaborator for over 17 years and codeveloped [28] the non-equilibrium extension of Wilson's numerical renormalization group which is the foundation of the scattering states approach to steady state currents applied in this paper. Furthermore, we had many fruitful discussions with Avi on the electron-phonon coupling and profited a lot from his exceptionally clear written paper [14]. We acknowledge financial support by the German-Israel Foundation through Grant No. 1035-36.14 and supercomputer support by the NIC, Forschungszentrum Jülich under project no. HHB000.

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